Progress report on measurement and fitting of pulse shapes of moderators at IPNS

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ABSTRACT

We present a progress report on measurements and fitting of pulse shapes for neutrons emerging from one solid and two liquid methane moderators in IPNS. A time-focused crystal spectrometer arrangement was used with a cooled Ge monochromator. Data analysis of one of the liquid methane moderators has shown the need for some generalization of the Ikeda-Carpenter function that worked well for fitting pulse shapes of polyethylene moderators. We describe attempts to model physical insight into the wavelength dependence of function parameters.

1. Introduction

At the time of this writing, the Intense Pulsed Neutron Source (IPNS) at Argonne National Laboratory is in its eighth year of successfully providing spectroscopists with slow (< 10eV) neutrons. The IPNS uses solid and liquid methane moderators to slow down fast (MeV) neutrons from a depleted uranium primary source. Fast neutrons are obtained by bombarding the depleted uranium target at 30 Hz with 80-ns bursts of 450 MeV protons. The fast neutrons are produced in pulses of the same frequency and approximate duration as the proton pulses. Thus, a convenient origin in time is established for neutron time-of-flight studies.

The neutrons emerge from the moderators in pulses also, but on quite a different time scale. The thermalization process in the moderators causes a substantial energy-dependent broadening of the neutron pulse which can amount to tens of microseconds. The end result is a fundamental limit of resolution established by the neutronic properties of the moderators. A quantitative understanding of the neutron emission-time distributions, or pulse shapes, for a moderator is necessary for neutron time-of-flight data analysis.

To this end, measurements were made of the neutron pulse shapes of the one solid and two liquid methane moderators in IPNS. This progress report briefly describes these measurements and the attempts made to fit mathematical functions to the data.

2. Measurements of Pulse Shapes

We measured the neutron pulse shapes of one solid and two heterogeneously poisoned liquid methane moderators. The solid methane "C" moderator measures 7.9 x 10.0 x 10.5 cm^3 in size, has 3.95-cm-deep grooves on the viewed face and can be cooled with liquid helium to temperatures as low as 10 K. The continuously-recirculated liquid methane "H" moderator measures 5 x 10 x 10 cm^3, is cooled to about 100 K, and is heterogeneously poisoned with a 0.5-mm Gd plate 2.5 cm below the viewed surface. The liquid methane "F" moderator has the same dimensions and temperature but is poisoned with two Gd plates 1.7 cm below its two viewed surfaces. The beamports at IPNS used to view the moderators for these measurements were C3, H1, and F1, respectively.

The experimental procedure follows closely that outlined by Ikeda and Carpenter when they made similar measurements of polyethylene moderators at IPNS.[1,2] The arrangement calls for a crystal monochromator and a detector with the correct geometrical orientation to achieve time focusing. That is, only reflections of neutrons having the wavelengths allowed by the Bragg diffraction equation will be seen by the detector. In addition, the experimental geometry assures that the neutrons of each particular order of reflection have flight path lengths and Bragg angles such that they reach the detector in exactly the same amount of time. This is true in spite of the fact that considerable changes of position of origin on the moderator, position and angle of the scatterer from the crystal, and positions of interactions in the detector are accepted in the measurements, which enhance the intensity. Fig. 1 shows a diagram of the experimental arrangement.

The conditions for time focusing in this arrangement are as follows:

$$P=L_f/L_i$$

$$\tan\theta_m = \tfrac{1}{2}(1+P)\cot\theta_B$$

$$\tan\theta_D = \tfrac{1}{2}(1+1/P)\cot\theta_B$$

$$\cot\theta_C = \frac{\cos\theta_D\tan\theta_m+\sin(2\theta_B+\theta_D)}{2\sin\theta_B\sin(\theta_B+\theta_D)} \qquad (1)$$

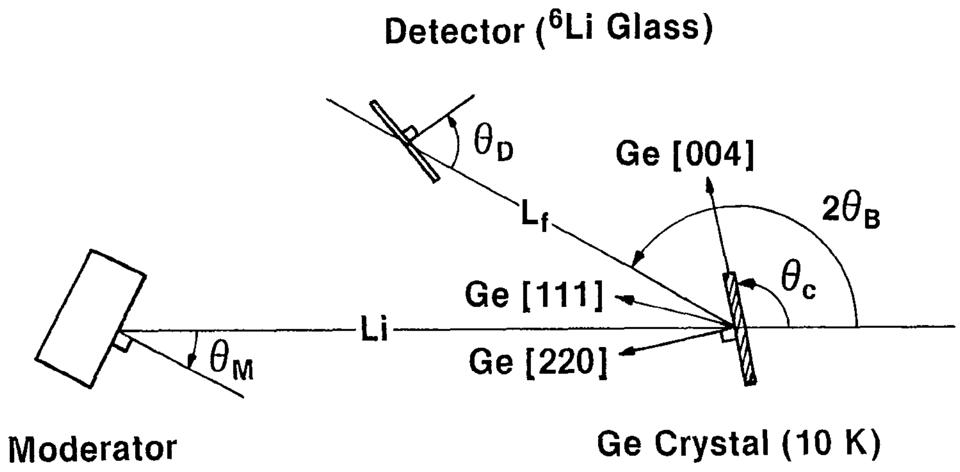

Fig. 1. Time-focused arrangement of moderator, monochromator, and detector used in these measurements.

The monochromator was a 1-mm thick, 25-mm diameter Ge crystal which had been cut parallel to (110) planes so that the (111) reflecting planes used were not parallel to the physical plane of the cut face. The Bragg angle was $\theta_B=60°$. Each of the beam ports used views its moderator at the angle $\theta_m=18°$ (focusing cannot be accomplished when $\theta_m=0°$ except when $\theta_B=90°$ (backscattering).) The detector was a 2-mm thick, 75-mm diameter ^{6}Li glass scintillator and was placed to intercept neutrons scattered at $2\theta_B=120°$. For each of the moderator measurements, the flight path lengths L_f and L_i were chosen so that their ratio P was equal to 0.1. For example, for the F moderator the crystal was positioned at a distance $L_i=13.44$ m from the moderator and the detector was positioned at a distance $L_f=1.34$ m from the crystal. This flight path ratio corresponds to a crystal angle, $\theta_C=95°$, and a detector angle, $\theta_D=73°$.

The Ge crystal was cooled to about 30 K to increase reflectivity and gain higher intensity for the high order reflections. The refrigeration and vacuum equipment used, however, did not maintain a constant temperature and there was warming during each of the data collection runs of at least 10 K.

From the neutron time-of-flight spectra obtained for the F moderator, good data was available through the ninth order reflection (206 meV) and reflections were observed through the 16th order reflection (650 meV). The statistics for these higher order reflections are poor. This will be addressed again in the analysis section. There are several factors contributing to to the low count rates for this measurement, including: the diminishing beam intensity for this long path length, and problems with cooling, vacuum, and geometrical alignment. Fig. 2 is a portion of the raw time-of-flight spectrum displaying six reflections. The Ge(444) reflection (1.41 Å) has the best statistics with nearly 1500 counts in the peak channel. Fig. 3 is an expanded view of the Ge(333) reflection (1.88 Å).

These high resolution measurements immediately reveal the very sharp rise in the leading edge. In the case of fig. 3, the 10-90% rise time is about 8 microseconds, a relative wavelength resolution of about 0.1% at this flight path length.

Spectra from the H moderator show pulses that look very similar (as expected, although not identical because of the different poisoning) to those of the F moderator. The exception is that because of better alignment, the peak integrals in the spectra from the H moderator are several times greater than those of the F moderator, so that the measurements from the H moderator are statistically more reliable.

Spectra from the solid methane C moderator show good peak statistics and clearly illustrate the presence of the physical

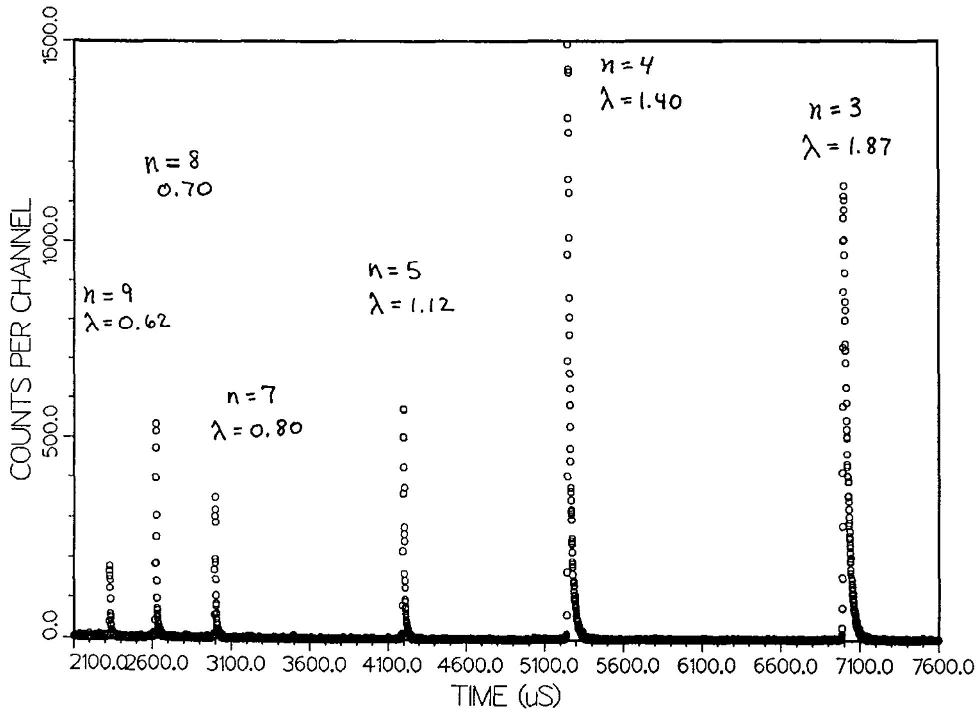

Fig. 2. Ge reflections observed in the F moderator measurement with L_i=13.4 m, L_f=1.34 m.

grooves in the moderator. Pulse shapes from this moderator are actually composed of two individual pulse shapes. The leading edge, or shoulder, represents the pulse of neutrons emerging from the top of the moderator "fins" and the second peak represents the pulse of neutrons of the same energy emerging from the bottom of the grooves. The second pulse is delayed by an amount of time equivalent to that required by the neutrons to travel the length of the grooves. Fig. 4 is a view of the Ge(444) reflection (1.41 A) and clearly shows that the intensity of neutrons from a grooved moderator is greatest from the base of the grooves.

3. Analysis and Discussion

To date, attempts to fit mathematical functions to the data

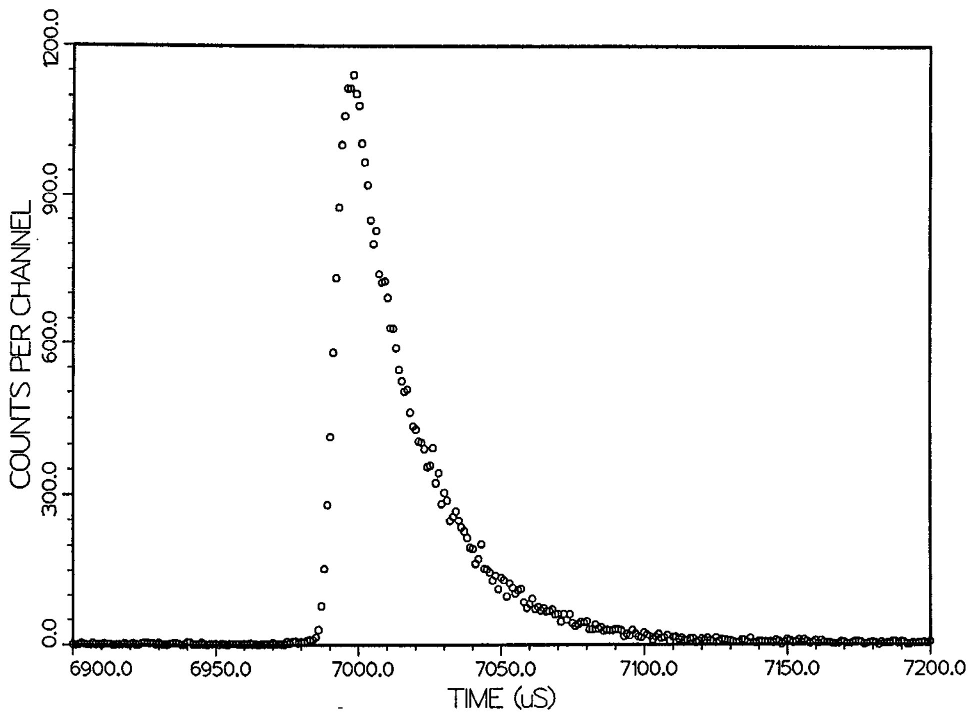

Fig. 3. Ge(3,3,3) reflection observed in F moderator measurement, 1.88 Å.

have been limited to the F moderator which, although having the least favorable counting statistics, were obtained first. Analysis of F moderator data was continued well after data had been collected from the similar H moderator. This may prove to have been unwise because several evolutionary steps have been made in the functions to provide better fits. These changes were based on results of analysis of the F moderator data without comparing function performance on the H moderator.

Ikeda and Carpenter[1] developed a function which fit the pulse shapes of polyethylene moderators extremely well. The function has four wavelength-dependent parameters and attempts to model some of the physical processes that occur in neutron thermalization. The function has the form

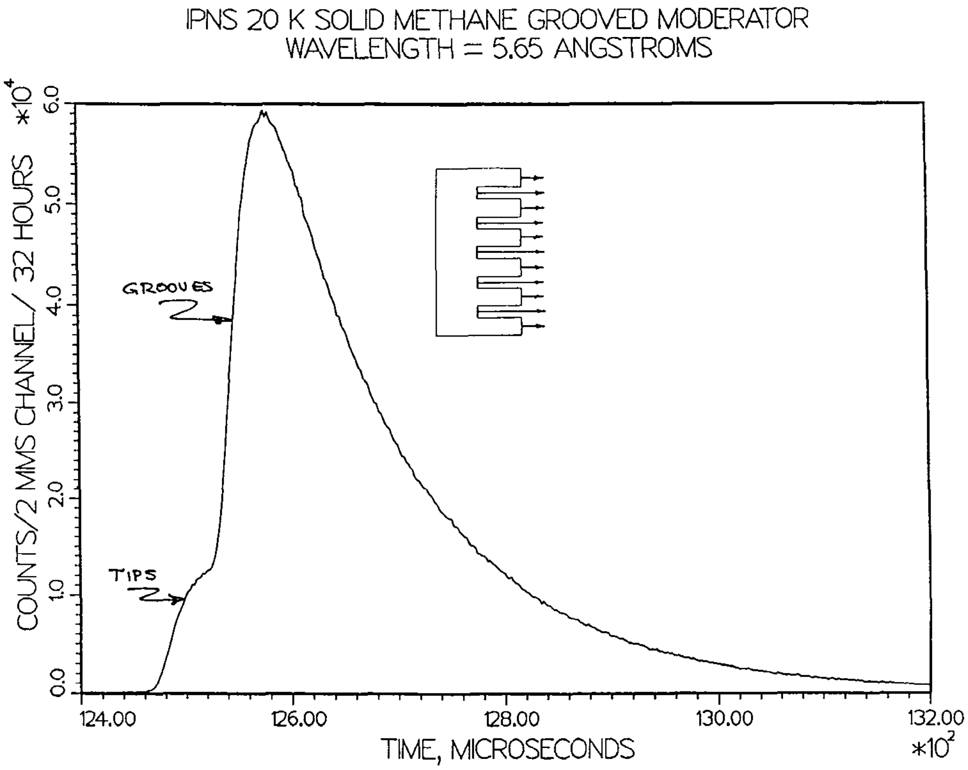

Fig. 4. Ge(4,4,4) reflection observed in C moderator measurement, 1.41 Å.

$$\Psi(a,\beta,R,t)=a/2\{(1-R)(at)2e{-}at + 2Ra2\beta/(a-\beta)3$$
$$\times\ [e^{-\beta t}-e^{-at}(1+(a-\beta)t+\tfrac{1}{2}(a-\beta)^{2}t^{2})]\} \qquad (2)$$

where, $a=v\Sigma_s$; the neutron velocity times the macroscopic scattering cross section.

R= fraction of the area of the peak in the 2nd term, the storage or thermalization term.

β= a time constant characterizing the decay of the storage term.

The function makes sense physically from the basics of neutron slowing-down theory. Neutrons emerging from a pulsed moderator

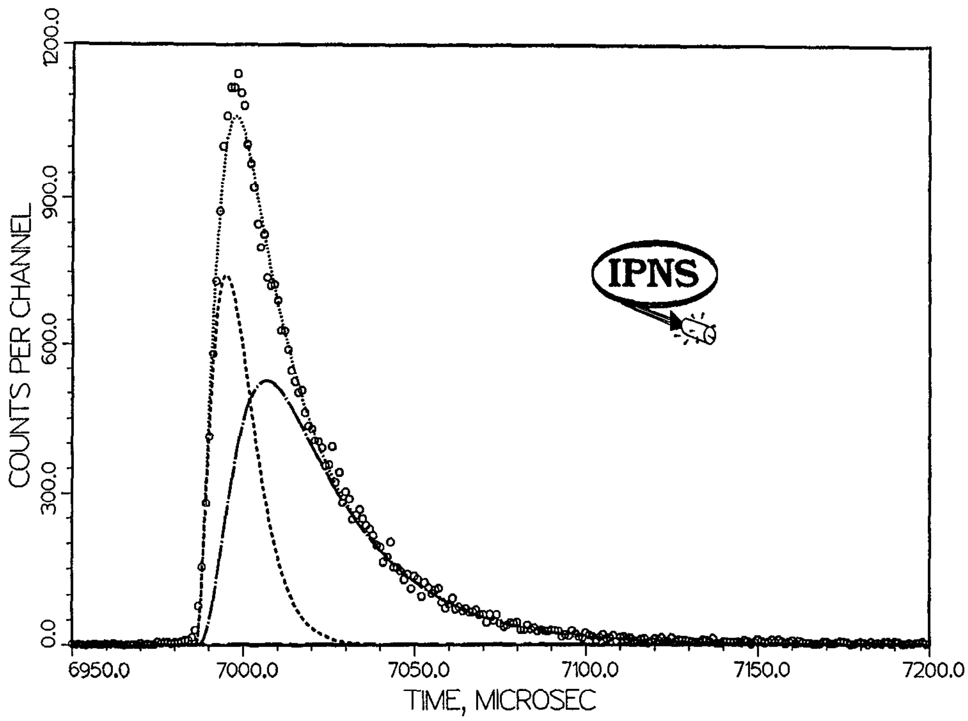

Fig. 5. Ge(3,3,3) reflection for F moderator. The dotted line is calculated by eq. (2) with parameters $\Sigma=2.88$ cm^{-1}, $\beta=0.04/\mu$sec, and R=0.65.

consist of two terms: a slowing-down component, representing neutrons that have emerged in the process of slowing down before thermalization, and a storage component, representing neutrons that have reached thermal equilibrium with the moderator and leak from it with a characteristic wavelength-independent decay constant β. The first term in eq. (2) represents the component of the pulse of a particular energy that is in the 1/E slowing-down spectrum (slowing-down term) and the second term (storage term) represents the component of the pulse that is in the Maxwellian part of the spectrum. The function is the sum of a delta function and the decaying exponential storage term convoluted with the slowing-down function.

Pulse shapes of the F moderator were fitted using this function and maintaining the three parameters described above, a scale factor, and a delay time t_0, as free parameters of wavelength. The results of the fitting, while encouraging, were not spectacular. Fig. 5 shows the results of fitting the Ikeda-Carpenter (IC) function to the data for the Ge(333) (1.88 Å) F moderator reflection. The figure indicates that the slowing-down term is important in fitting the rising edge of the peak and the storage term is dominant in the decaying tail of the pulse shape for neutrons of this wavelength. Characteristic of these fits was a systematic underestimation of the height and integral of each peak. This experience suggested that some modification was needed in the IC function.

The slowing-down term in the IC function is the simple description of neutrons slowing down in an infinite medium of free protons (the "proton gas" model). We suggest that a generalization in the exponent in the slowing-down term from 2 be made to account for some of the molecular characteristics of thermalization. Such a generalization is plausible on the grounds of using the Sachs-Teller effective mass[3] to approximate the scatterers in a liquid, where molecular effects can be important, with gas atoms of mass greater than the physical mass.

The function maintains the form of its predecessor but is more general in that it contains the incomplete gamma function. The generalized IC function has the form

$$\Psi(a,\beta,\nu,R,t) = (1-R)\mathfrak{F}(a,\nu,t) + RF(a,\beta,\nu,t) \tag{3}$$

where: $$F(a,\beta,\nu,t) = [\beta a^{\nu+1}/\Gamma(\nu+1)](a-\beta)^{-(\nu+1)}e^{-\beta t} \times \gamma((\nu+1),(a-\beta)t) \tag{4}$$

is the slowing-down term and,

$$\eth(a,\nu,t) = [a/\Gamma(\nu+1)](at)^{\nu}e^{-at} \quad (5)$$

is the storage term.

Again, pulse shapes of the F moderator were fitted. The generalized IC function was used while maintaining the parameters a, ß, ν, R, a scale factor, and t_0 as free parameters of wavelength. The results showed improvement over previous fitting in that discrepancies between best fits of the function to the data and the peak integral were eliminated. An example of a fit using the generalized IC function for the Ge(333) F moderator reflection with no restraint on the fitting parameters is shown in fig. 6. Allowing freedom of the parameters during the

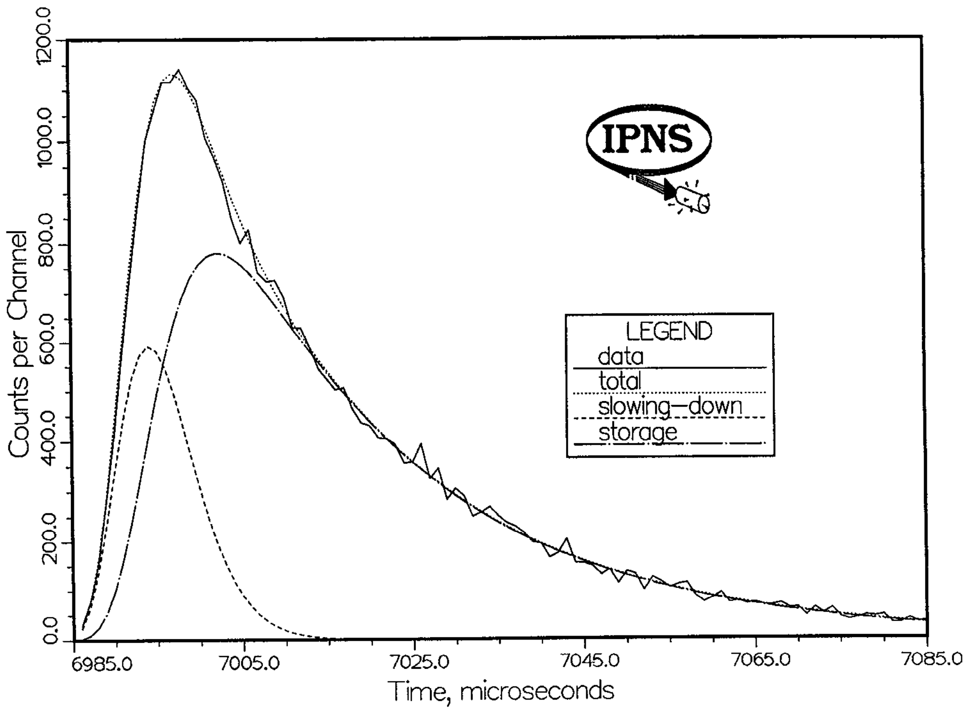

Fig. 6. Ge(3,3,3) reflection for F moderator. The dotted line is calculated by eq. (3) with parameters ν=7.4, Σ=2.88 cm^{-1}, ß=0.041/µsec, and R=0.81. The estimated chi-square value for this fit is 1.288.

fitting, however, produced some unexpected variations as a function of wavelength (energy). Most disturbing was the large fluctuation in the parameter β. This parameter represents the decay constant of the storage term which, to first approximation from slowing-down theory, should be representative of the fundamental eigenvalue of the Maxwellian and should be independent of energy.[4]

So as not to completely disregard the theory, the parameter β was "fixed" for all energies and fits were made again with the remaining four parameters maintained as free parameters for each wavelength. The results of this trial showed an interdependence of the parameters a, ν, and t_0. Fits of pulse shapes of a particular wavelength of almost equal quality (based on an estimation of the chi-square values for the fits; the least-squares iteration was not carried all the way to the minimum of χ^2) could be obtained with different triplet sets of a, ν, and t_0. A three-dimensional linear regression analysis for these parameters was performed for one of the peaks, which demonstrated their linear interdependence within a rather wide range of values of a, ν, and t_0 . Rather than perform this task for each of the peaks, we chose to include a wavelength-dependent functional form for each of the parameters in the fitting routine and fit all the pulse shapes from the moderator data simultaneously.

The functional forms for the four wavelength-dependent parameters (β was a fitted parameter, but constant at all wavelengths) were chosen with the desire to implement a little knowledge of physics and provide for some physically believable parameters. The wavelength dependent functions were now of the form

$$t_0 = A + B\lambda, \tag{6}$$

$$a = v\Sigma_s, \tag{7}$$

where v= neutron velocity, and

Σ_s= macroscopic scattering cross section

$$\Sigma_s = (S_1{}^2 + S_2{}^2\lambda^2)^{1/2}, \tag{7'}$$

$$R = \exp(-E/E_0), \text{ and} \tag{8}$$

$$\nu = 2. + c^2\exp(-E/E') \tag{9}$$

This representation of the generalized IC function now called for fitting seven new wavelength-independent parameters to determine the four wavelength-dependent parameters for each peak and an eighth parameter for the wavelength-independent parameter ß as before. Each peak was allowed its own amplitude factor. The time delay t_0 shown in eq. (6) represents the flight time from moderator to detector plus a fixed shift of the time origin and is necessarily linear in form. The functional forms chosen for Σ_s and R have proven successful earlier.[1] The form for the exponent parameter ν was chosen with the feeling that it should approach a lower limit of 2 for shorter wavelengths and should be in some fashion greater than two for longer wavelengths to account for the increasing Sachs-Teller effective mass.

Fits to the F moderator data using these functions presented another problem. The function was not adequately describing the decaying tail of the higher-order reflections; it was dying away too rapidly, underestimating the tails of the short wavelength pulse shapes. One explanation was the possibility of the existence of another decay constant corresponding to a higher-order energy or spatial eigenvalue of the moderator. (When ß was a free parameter, best fits were obtained with different decay constants for the higher-order reflections.) Another possibility was the need for a better representation of the parameter R than the simple Boltzmann function of eq. (8). The latter alternative was pursued by including the energy spectrum relationship of the slowing-down and storage terms of the generalized IC function. That is, representing the energy spectrum as a Maxwellian component smoothly joining with the 1/E slowing down component in the epithermal region. Here, use was made of a generalization of Westcott's[5] joining function

$$\Delta(E) = [1+(E_{co}/E)^{s}]^{-1}. \tag{10}$$

where: E_{co}= a cutoff energy corresponding to about $5k_BT$ and,

s= a parameter suggested by Westcott to be about seven.

The generalized IC function could then be written as

$$\Psi(a,\beta,\nu,\Delta,E_T,E,A,t) = (\Delta/E)\Phi(a,\nu,t)+A^2e^{-E/E}\,EF(a,\beta,\nu,t) \tag{11}$$

where F and Φ have their previous meanings and A^2 is another amplitude factor.

The function did not perform as well as had been hoped on the F moderator data. In some respects it performed even worse, underestimating the peak integrals for the intermediate wavelength pulse shapes.

Shortly before the time of this writing a second decay constant (also independent of energy) was added. This created an entirely new storage term corresponding to an additional moderator eigenvalue. The function describing the wavelength dependence of R was dropped and in its place, free independent coefficients for each of the three terms were incorporated. The new function has the form

$$\Psi(a,\nu,\beta_1,\beta_2,A_1,A_2,A_3,t)=A_1\Phi(a,\nu,t)+A_2F(a,\beta_1,\nu,t)+A_3F(a,\beta_2,\nu,t) \quad (12)$$

This pulse shape function provides the best fits to the F moderator data using simultaneous, multi-peak fitting of all attempts to date. A plot of the Ge(333) pulse shape fit with this function is shown in fig. 7; this does not produce as good a fit as when peaks were fit individually as was shown in fig 6.

4. Concluding Remarks

Pulse shapes have been measured of the one solid and two liquid methane moderators at IPNS. Attempts to analyze data from the liquid methane F moderator have suggested the need for generalization of the Ikeda-Carpenter function and the inclusion of physical models of parameter wavelength dependence in the fitting process. Results may also indicate the need to include an additional decay constant for an additional moderator eigenvalue other than that of the fundamental mode.

The counting statistics for the F moderator are not of the same quality or as those obtained the H and C moderators. Future work will involve using the experience and lessons learned from analysis of the F data to complete the analysis of the remaining moderators.

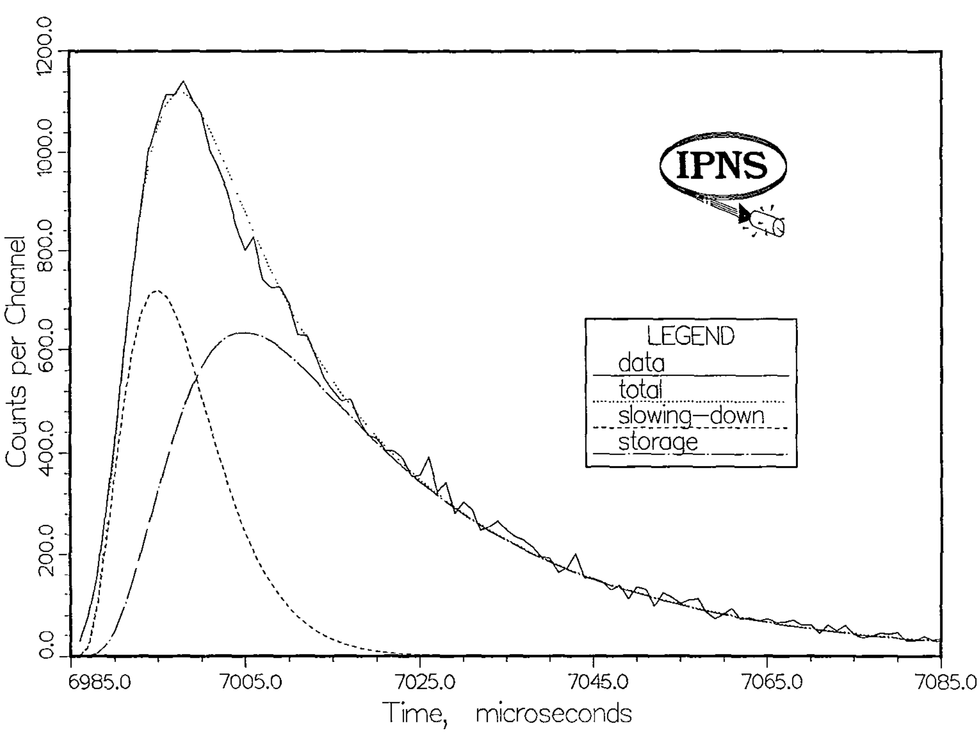

Fig. 7. Ge(3,3,3) reflection for F moderator. The dotted line is calculated by eq. (12) with parameters ν=2.88, Σ=1.53 cm^{-1}, β_1=0.042/μsec, β_2=0.036/μsec, A_1=0.282, A_2=0.619, A_3=0.099. The estimated chi-square value for this fit is 1.802. Parameters for this fit were calculated by simultaneously fitting reflections in the F moderator data (see text). The storage term shown is the sum of two individual storage terms with two decay constants.

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